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A. Wright, A. Hannon, A. Clare, R. Sinclair, W. Johnson, et al.. A NEUTRON DIFFRACTION INVESTIGATION OF THE ATOMIC AND MAGNETIC STRUCTURE OF AMORPHOUS Dy₇Ni₃. Journal de Physique Colloques, 1985, 46 (C8), pp.C8-299-C8-303. 10.1051/jphyscol:1985844 . jpa-00225187

HAL Id: jpa-00225187

<https://hal.science/jpa-00225187>

Submitted on 4 Feb 2008

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A NEUTRON DIFFRACTION INVESTIGATION OF THE ATOMIC AND MAGNETIC
STRUCTURE OF AMORPHOUS Dy_7Ni_3

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Résumé - La structure atomique et magnétique de Dy_7Ni_3 amorphe a été étudiée en diffraction neutronique à partir d'une méthode de double substitution isotopique qui s'est avérée extrêmement puissante.

Abstract - The atomic and magnetic structure of melt-spun amorphous Dy_7Ni_3 is being studied using the neutron diffraction double-null isotopic substitution technique. It is concluded that the double-null technique is a very powerful method for studying the atomic and magnetic structure of appropriate binary amorphous alloys.

1 - INTRODUCTION

Amorphous solids are of two main types, those with a structure based on a covalent network, with a well defined structural unit, and those such as amorphous metallic alloys whose structure is much nearer a random close packing of spheres. The less well defined structure of the latter means these materials are in general even less understood than their covalent counterparts, despite the fact that amorphous metallic alloys are of considerable academic and commercial interest, particularly in respect of their magnetic properties. Work is therefore in progress to investigate in detail one particular alloy (melt-spun amorphous Dy_7Ni_3), chosen because of its special neutron scattering properties, using not only the full range of neutron scattering techniques but also X-ray diffraction and bulk thermodynamic and magnetic measurements. Preliminary results are presented here from a neutron diffraction study of the ambient atomic structure of amorphous Dy_7Ni_3 and of the magnetic structure as a function of temperature in the range 7-293K.

The structure of an amorphous material lacks periodicity, symmetry and long range order and is usually isotropic on a macroscopic scale. This leads to a structural description in terms of a one-dimensional real space correlation function. For a sample containing n elements there are $n(n+1)/2$ independent component (partial) correlation functions,

$$d_{jk}(r) = 4\pi r [\rho_{jk}(r) - \rho_k^0] \quad (1)$$

each of which gives rise to an equivalent interference function,

$$Q_{ijk}(Q) = \overline{b_j b_k} \int_0^\infty d_{jk}(r) \sin Qr dr \quad (2)$$

in reciprocal (intensity) space/1.2/. A single neutron diffraction experiment, however, measures only a linear sum of these components

$$Q(Q) = \sum_j \sum_k Q_{ijk}(Q) \quad (3)$$

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in which the j summation is taken over the atoms in one composition unit and that for k over atom types. An important advance in recent years has been the development of experimental techniques to separate experimentally the component interference functions for multi-element samples and hence to determine the corresponding correlation functions. In the case of neutron diffraction this may be achieved for certain favourable systems by means of magnetic diffraction experiments /3/, the anomalous dispersion technique /4,5/ or isotopic substitution /6/.

Isotopic substitution is based on the fact that in general different isotopes of a given element have different neutron scattering lengths \bar{b} and hence the average value for a given element A can be adjusted by varying the isotopic composition. The main problem with isotopic substitution is that the differences in scattering length achieved are frequently small and the scattering lengths themselves are often not known very accurately. Thus errors can rapidly build up, as shown by the large uncertainties in many published component correlation functions. The potential scattering of a neutron by a nucleus is normally accompanied by a phase change of π , but for a small number of isotopes the scattered wave is in phase with the incident wave leading to a negative scattering length. A few elements (H, Li, Ti, Cr, Ni, Sm, Dy, W) can therefore be obtained as isotopes with both positive and negative scattering lengths and, since the scattering length for element A in a given sample is simply the isotopically averaged value, this allows the special possibility of $\bar{b}_A=0$, which is the basis of the null technique /7/.

If one of the elements in a binary alloy has a zero scattering length then, as may be seen from equ. (2) and (3), a measurement of the interference function yields the like-atom interference function for the other element directly. The null technique is particularly powerful for binary systems, however, when both elements can be obtained with a zero scattering length as in the case of amorphous Dy_7Ni_3 . Both like-atom interference functions can then be obtained by direct measurement and the unlike-atom interference function by simply subtracting the other two from that for a sample containing the natural elements.

The fact that both Dy and Ni can be obtained with a zero scattering length also means that it is possible to prepare a sample for which the nuclear scattering is completely incoherent which may be used both for magnetic diffraction and inelastic scattering experiments. The ambient temperature diffraction pattern from this sample yields the magnetic form factor, without interference from coherent nuclear scattering, while at lower temperatures the sample exhibits magnetic ordering. The magnetic structure may therefore be determined from the difference between diffraction patterns measured above and below the macroscopic transition temperature T_c . For a sample which scatters only incoherently the density of vibrational states $g(\omega)$ can be accurately obtained from an inelastic neutron scattering experiment without the need to invoke the incoherent approximation.

II - EXPERIMENTAL DETAILS AND DISCUSSION

The composition Dy_7Ni_3 is close to the eutectic composition and hence the samples could be prepared by melt-spinning. Four samples were prepared:

- (i) Dy_7Ni_3
- (ii) 0Dy_7Ni_3
- (iii) $Dy_7{}^0Ni_3$
- (iv) ${}^0Dy_7{}^0Ni_3$

In which a leading superscript zero is used to denote a zero neutron scattering length. Ambient temperature diffraction patterns were obtained for all four samples using the D2 and D4 diffractometers at I.L.L., those from D2 being given in fig. 1. All the diffraction patterns exhibit a general fall-off with increasing scattering vector, Q , due mainly to the Dy magnetic form factor as discussed below. The great similarity between the data for Dy_7Ni_3 and $Dy_7{}^0Ni_3$ shows that the scattering from the former is dominated by the Dy-Dy component. (The latter is sensitive only to the Dy-Dy correlations). The differences between the two patterns are mainly due to the

Dy-Ni contribution and are most apparent around the second and third peaks. The ${}^0\text{Dy}_7\text{Ni}_3$ diffraction pattern (Ni-Ni correlations) is very different in that the distinct scattering contribution is very small. There are several reasons for this. First, the neutron scattering length of natural Ni is only $1.03 \times 10^{-14}\text{m}$ compared to $1.69 \times 10^{-14}\text{m}$ for natural Dy /8/ and second, the atomic fraction of Ni in the samples is less than half that of Dy. Also since Ni is the smaller atom it is to be expected that the Ni-Ni first co-ordination shell will be rather poorly defined relative to those for Dy-Ni and Dy-Dy correlations.

The shape of the Dy magnetic form factor can most easily be seen from the ${}^0\text{Dy}_7{}^0\text{Ni}_3$ diffraction pattern. The fact that this pattern shows virtually no structure confirms a paramagnetic state at room temperature. (The slight feature in the region of the first peak for $\text{Dy}_7{}^0\text{Ni}_3$ is almost certainly due to a very small residual Dy scattering length). The ${}^0\text{Dy}_7{}^0\text{Ni}_3$ sample was used to investigate the magnetic structure at low temperatures by measuring the diffraction pattern at 7, 44 and 96K. The first magnetic diffraction peak coincides with that for the $\text{Dy}_7{}^0\text{Ni}_3$ sample indicating that the magnetic ordering is ferromagnetic. The temperature dependence of this peak was investigated in more detail by performing additional short scans at 20, 33, 38, 53, 64 and 148K and these are shown together with the relevant portions of the full scans in fig.2. after subtraction of the ambient temperature pattern to give magnetic distinct scattering $i^M(Q)$ /3/.

There is structure in $i^M(Q)$ to at least 148K showing that remnant short range magnetic ordering persists well above the macroscopic transition temperature of $38 \pm 1\text{K}$. The variation in the extent of short range magnetic ordering with temperature can be more easily seen from fig.3 in which the height of the first peak in $i^M(Q)$ is plotted as a function of temperature. This shows that the short range magnetic ordering falls off smoothly with temperature with no apparent discontinuity at T_c .

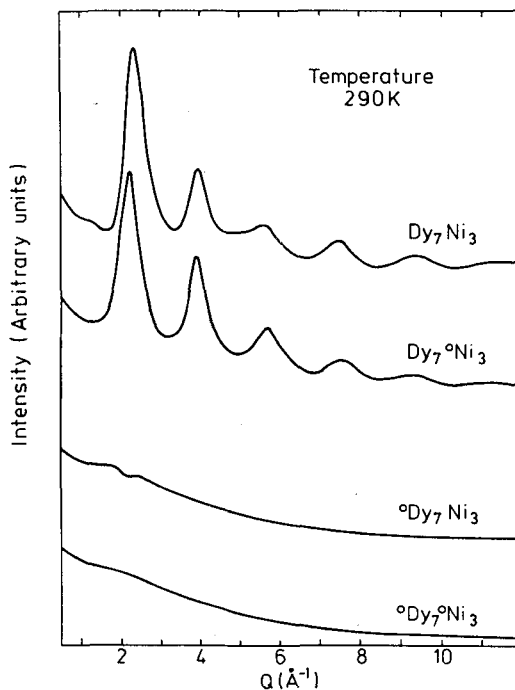


Fig. 1 - Diffraction patterns for melt-spun amorphous Dy_7Ni_3 .

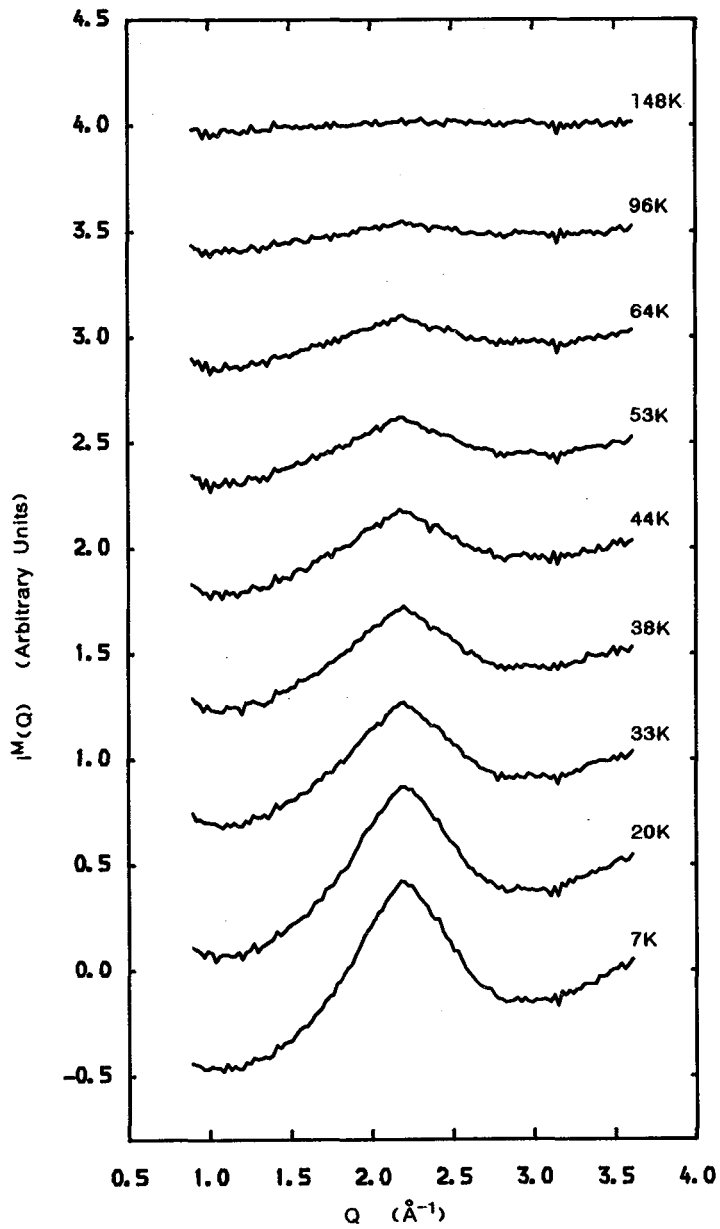


Fig.2 - The magnetic scattering $i^M(Q)$ as a function of temperature.

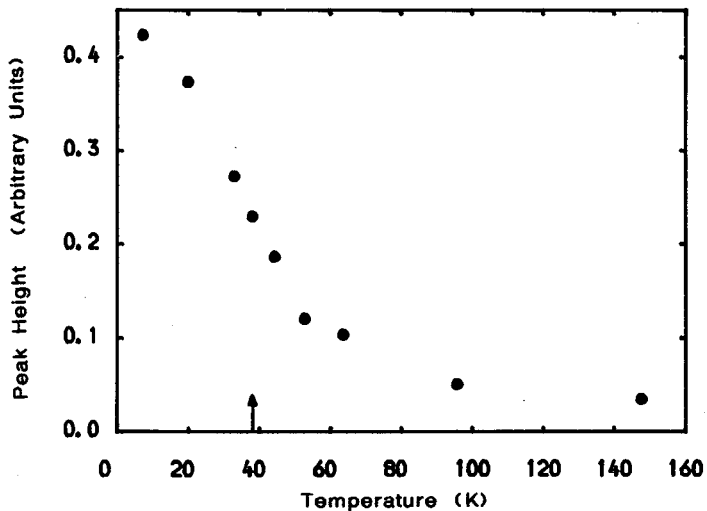


Fig. 3 - The temperature dependence of the height of the first peak in $I^M(Q)$. The vertical arrow indicates T_c ($38 \pm 1K$).

III - CONCLUSION

From the present series of experiments it is apparent that in suitable systems the double-null isotopic substitution technique is an extremely powerful method for studying the atomic and magnetic structure of a binary amorphous alloy. Further small Q (neutron magnetic critical scattering) and inelastic neutron scattering measurements are planned and these will be reported elsewhere together with a detailed analysis of the above diffraction data.

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